METHANE EMISSIONS OF DIFFERENTLY FED DAIRY COWS AND CORRESPONDING METHANE AND NITROGEN EMISSIONS FROM THEIR MANURE DURING STORAGE

D. R. KÜLLING[1], FRIGGA DOHME[2], H. MENZI[1,3], F. SUTTER[2,4], P. LISCHER[5] and M. KREUZER[2*]

[1] *Federal Research Station of Agroecology and Agriculture (FAL), Liebefeld, Berne, Switzerland;* [2] *Institute of Animal Sciences, Animal Nutrition, Swiss Federal Institute of Technology Zurich (ETH), ETH Zentrum/LFW, CH-8092 Zurich, Switzerland;* [3] *Present affiliation: Swiss College of Agriculture, Zollikofen, Switzerland;* [4] *Present affiliation: Swiss Centre for Agricultural Extension (LBL), Lindau, Switzerland;* [5] *Constat, Ahornweg 46, Spiegel, Switzerland*
(author for correspondence, e-mail: michael.kreuzer@inw.agrl.ethz.ch)*

(Received 19 March 2001; accepted 2 November 2001)

Abstract. This study investigated the effects of supplementing 40 g lauric acid (C_{12}) kg^{-1} dry matter (DM) in feed on methane emissions from early-lactating dairy cows and the associated effects on methane, nitrous oxide and ammonia release from the manure during storage. Stearic acid (C_{18}), a fatty acid without assumed methane-suppressing potential in the digestive tract of ruminants, was added at 40 g kg^{-1} DM to a control diet. The complete feed consisted of forage and concentrate in a ratio of 1.5:1 (DM basis). The manure was stored for 14 weeks either as complete slurry or, separately, as urine-rich slurry and farmyard manure representing two common storage systems. Methane release of the cows, as measured in respiratory chambers, was lower with C_{12} by about 20%, but this was mostly resulting from a reduced feed intake and, partly, from a lower rate of fibre digestion. As milk yield declined less than feed intake, methane emission per kg of milk was significantly lower with C_{12} (11.4 g) than with C_{18} (14.0 g). Faeces of C_{12}-fed cows had a higher proportion of undigested fibre and accordingly methane release from their manure was higher compared with the manure obtained from the C_{18}-fed cows. Overall, manure-derived methane accounted for 8.2% and 15.4% of total methane after 7 and 14 weeks of storage, respectively. The evolution of methane widely differed between manure types and dietary treatments, with a retarded onset of release in complete slurry particularly in the C_{12} treatment. Emissions of nitrous oxide were lower in the manures from the C_{12} treatment. This partially compensated for the higher methane release from the C_{12} manure with respect to the greenhouse gas potential. The total greenhouse gas potential (cow and manure together) accounted for 8.7 and 10.5 kg equivalents of CO_2 cow^{-1} d^{-1} with C_{12} and C_{18}, respectively. At unaffected urine-N proportion ammonia and total nitrogen losses from stored manure were lower with C_{12} than with C_{18} corresponding to the differences in feed and nitrogen intake. The present results suggest that manure storage significantly contributes to total methane emission from dairy husbandry, and that the identification of effective dietary mitigation strategies has to consider both the digestive tract of the animals and the corresponding manure.

Keywords: ammonia, dairy cows, lipids, manure storage, methane, nitrous oxide

1. Introduction

In the aftermath of the United Nation's environmental conference in Rio 1992, political concern about the emissions of greenhouse gases such as carbon dioxide

Environmental Monitoring and Assessment **79:** 129–150, 2002.

(CO_2), methane (CH_4) and nitrous oxide (N_2O) gave rise to research for reduction potentials. Within the past 200 years, the atmospheric concentration of CH_4 and N_2O has increased by 115 and 15%, respectively (IPCC, 1996). Worldwide, agriculture is responsible for 35% of greenhouse gas emissions (IPCC, 1996), and domestic livestock is estimated to produce 80 Tg a^{-1} of CH_4 with another 25 Tg a^{-1} being emitted from their manure (Chynoweth, 1996). The annual greenhouse gas budget in Switzerland accounted for the equivalent of 0.046 Tg CO_2, of which 10% were CH_4 and 8% N_2O (Schmid *et al.*, 2000). Ruminants are responsible for a major part of CH_4 release, particularly in grassland dominated regions where ruminants are the most important natural converters of fibrous biomass to valuable protein for human nutrition (Immig, 1996). Similar as for rice paddies (Agnihotri *et al.*, 1999), mitigation strategies for ruminant livestock are intensively under investigation. Van Nevel and Demeyer (1996) as well as Moss *et al.* (2000) reviewed the current knowledge on effective nutritional means to suppress CH_4. As one promising natural option the use of lipids was described, and medium-chain fatty acids (Dohme *et al.*, 2001) and fats thereof (Machmüller and Kreuzer, 1999; Dohme *et al.*, 2000) turned out to be particularly efficient against ruminal CH_4 synthesis. It is, however, unclear, whether or not CH_4 mitigation strategies, which are effective in the enteric fermentation of the animal, are prone to a compensatorily higher CH_4 release from the manure during storage, as the amount of residual fermentable organic matter available may be higher. Hashimoto *et al.* (1981) and Külling *et al.* (2001) described in cattle that certain diets had an effect on manure CH_4 production. The CH_4 from stored manure as a proportion of total CH_4 from cattle was estimated to range from 13 to 14% (Anonymous, 1996; Minonzio *et al.*, 1998). However, there are still principal problems in the estimation of CH_4 emission from livestock manure (Johnson and Ward, 1996) and, to our knowledge, a simultaneous comparison of CH_4 derived from the animal and their manure has not yet been performed under controlled and varying conditions.

Further importance for the overall estimation of the greenhouse gas potential of ruminant systems is given by the level of emission of nitrogenous trace gases from manure (Sommer *et al.*, 2000). Both N_2O, directly contributing to the global warming potential, and ammonia (NH_3), possibly inhibiting CH_4 release from manure (Hansen *et al.*, 1998), are important in this respect. So far only few investigators (e.g. Amon *et al.*, 1998; Külling *et al.*, 2001, 2002) have followed the emission of both greenhouse gases and NH_3 during several weeks of manure storage and it seems that the system and the duration of storage play an important and interactive role for the overall release of greenhouse gases.

The objectives of the present study were (i) to determine the proportion of CH_4 emitted from the rumen and during manure storage; (ii) to monitor the effects of replacing a long-chain saturated fatty acid (stearic acid; C_{18}) by a medium-chain saturated fatty acid (lauric acid, C_{12}) known as one of the fatty acids effective against ruminal CH_4 release *in vitro* (Dohme *et al.*, 2001); (iii) to assess the effect of system (complete slurry vs urine-rich slurry/farmyard manure) and duration (7 vs 14 weeks) of manure storage for the overall release of CH_4. In order to identify interactions of CH_4 formation and the emission of other trace gases, N_2O and NH_3 release from manure were also followed.

2. Materials and Methods

2.1. Animal experiment

Twelve early-lactating Brown Swiss cows (31.3±5.1 kg milk d^{-1} with 4.17±0.39% fat and 3.14±0.15% protein) weighing on average 635±56 kg were subjected to two dietary treatments in two subsequent series of three cows per each treatment. The treatments differed in the type of fatty acid, either lauric acid (C_{12}) or stearic acid (C_{18}), both purchased from Cognis Deutschland GmbH (Düsseldorf, Germany) and supplemented at a level of 40 g kg^{-1} feed dry matter (DM) equivalent to approximately one third of all fatty acids in the feed (Table I). The complete diets were composed of forage and concentrate in a ratio of 3:2 on a DM basis. The forage ration consisted of a mixture of young rye-grass dominated grass silage, maize silage and hay harvested at a late growth stage from a permanent meadow highly diverse in species composition. The barley-based concentrate (Table I) was provided separately from the forage. The supply of absorbable protein and net energy was calculated according to RAP (1999). Feed was allocated in a restricted manner which was oriented towards the individual requirements of the cows for maintenance and milk yield as defined by RAP (1999).

Each animal performed a 10-day control period on the C_{18} diet and a 15-day main period receiving either the C_{12} or the C_{18} diet. During the last six days of the main period faeces and urine were quantitatively collected. Additionally, gaseous exchange of the cows was measured in open-circuit respiratory chambers as described by Sutter and Beever (2000) on days three and four of the collection period. The animals were tethered in individual stalls complete with slatted floor fitted for quantitative collection of faeces and urine. These were similar in the cow-shed and in the respiratory chambers. Urine was separated from the faeces by urinals attached with Velcro straps glued onto the clipped skin with special adhesive (Cyanolit 202, 3M AG, Rüschlikon, Switzerland). A small proportion of urine was acidified with 5 M sulphuric acid for determination of the initial urine N content. Excreta were sampled daily and stored separately at 4 °C. The two respiratory chambers, fitted also for milking twice daily, were air conditioned (relative humidity 59.3±1.05% (mean ± SD), air temperature 17.3±0.42 °C, air flow 27.2±0.71 m^3 hr^{-1}). Methane concentrations of the air flowing into the chambers (internal volume: 20 m^3) were measured for 0.3 min every 20 min while outgoing air was analysed for CH_4 every 1.3 min with an infrared analyser (Binos 1001, Leybold-Heraeus, Zurich, Switzerland). The air volume leaving the chambers was continuously recorded with in-line electronic flow meters (Swingwirl DV 630;

TABLE I
Composition of the diets as offered and as consumed in the animal experiment

Dietary treatment	Lauric acid (C_{12})		Stearic acid (C_{18})	
	As offered	As consumed	As offered	As consumed
Ingredients (g kg^{-1} dry matter (DM))				
Forage	600	710	600	600
Concentrate	400	290	400	400
Forage ingredients (g kg^{-1} DM)				
Grass silage	510		510	
Maize silage	320		320	
Hay	170		170	
Concentrate ingredients (g kg^{-1} DM)				
C_{12}	100		–	
C_{18}	–		100	
Barley	660		660	
Maize gluten	140		140	
Molasses	70		70	
Mineral and vitamin premix[a]	30		30	
Analysed composition				
Organic matter (g kg^{-1} DM)	920	914	919	919
Total fatty acids (g kg^{-1} DM)[b]	63	52	56	56
C_{12} (g kg^{-1} total fatty acids)	328	241	11	11
C_{18} (g kg^{-1} total fatty acids)	19	21	346	346
Neutral detergent fibre (g kg^{-1} DM)	329	358	320	320
Nitrogen (g kg^{-1} DM)	25.0	24.3	25.3	25.3
Absorbable protein at intestine (g kg^{-1} DM)[c]	100	95	101	101
Net energy for lactation (MJ kg^{-1} DM)[c]	7.3	6.9	7.2	7.2

[a] Supplied per kg concentrate: 5.1 g Ca, 1.5 g P, 1.2 g Mg, 1.2 g Na, 30 000 IU vitamin A, 6 000 IU vitamin D_3, 45 mg vitamin E.
[b] Calculated as triglyceride equivalents.
[c] According to RAP (1999).

Flowtec AG, Reinach, Switzerland). The chamber system was manually calibrated in advance of the CH_4 measurements, and automatic calibrations were performed every 3 hr including one calibration at the end of the 48 hr period. Calibrations lasted for about 9 min each. The experiment was conducted in accordance with the Swiss guidelines for animal welfare.

2.2. Manure storage experiment

Two typical farm manure collection and storage systems were simulated, the slurry system as applied in the buildings with slatted floors (tied or loose housing systems) collecting all excreta in the slurry, and the urine-rich slurry/farmyard manure system as typically applied in the traditional tied housing systems (Menzi *et al.*, 1997). In the latter system faeces are largely retained by straw and added twice daily to a manure stack outside the building, while the urine largely flows into an extra pit. The manure storage experiment was carried out as described in detail in Külling *et al.* (2001). In the present experiment, only faeces and urine collected in the first series of the animal experiment were employed, i.e. samples simultaneously obtained from three cows per treatment. These samples were homogenised and unified to one sample per treatment. In each experimental manure type 5 kg of excreta were filled into open plastic buckets. Complete slurry consisted of the actually excreted proportions of faeces and urine. Following the assumptions of Walther *et al.* (1994) for Swiss farm practice conditions, urine-rich slurry contained 90% of urine and 10% of faeces and the remainder formed the farmyard manure. Both slurries were mixed and diluted with 2.5 kg (deionised) water. Farmyard manure was supplemented with 0.23 kg straw (chopped to 5.1±0.2 cm length) corresponding to the typically added 1.75 kg straw cow^{-1} d^{-1} (Walther *et al.*, 1994). The complete farmyard manure samples were compacted with a weight of 2 kg cm^{-2}. The storage period lasted for 14 weeks. The slurries were held at 20 °C and 70% ambient humidity in a 40 m^3 sized laboratory ventilated at 0.33 m^3 s^{-1}. The farmyard manure containing buckets were kept in a heated bath at temperatures typical for stacked manure exposed to self heating as was determined in a preliminary investigation (Külling *et al.*, 2001). Accordingly, the initial temperature was set to 41 °C and then was gradually reduced by 2 °C per week. The two dietary treatments were tested in four replicates for each of the three manure types (total n = 24).

As suggested by Mosier (1989), a closed chamber was used to monitor CH_4 and N_2O emission rates, and NH_3 release was measured with a dynamic chamber. Chambers and their operation are described in detail in Külling *et al.* (2001). The buckets containing the manure samples and the covering buckets of the dynamic chambers had a height of 26 cm, an inner diameter of approximately 24 cm and a volume of 11.5 L. The manure samples used about 8 L and had a surface of 0.05 m^2, equivalent to 0.01 m^2 kg^{-1} undiluted manure. The volume of the closed chamber used for the determination of CH_4 and N_2O emission rates was variable from 11.5 to 15.5 L, thus allowing the repeated extraction of air samples (three samples per bucket and measurement date) without interrupting the accumulation. Methane in air samples was analysed with gas chromatography (CH_4/Non-CH_4 Carbohydrogen Analyser 55C, Thermo Environmental Instruments Inc., Franklin, MA, U.S.A.) and N_2O with diode laser spectroscopy (TDLAS 004, Aerodyne, U.S.A.). Ammonia measurements, carried out once a week per bucket in the beginning and every other week later on, were performed every 10 s during 30 min after covering the manure storage buckets by the dynamic chamber equipped with two ventilators (wind speed, 0.2 and 1.2 m s^{-1} at the inlet and the outlet, respectively). The final NH_3 values represented steady state and were used for further calculations. Outflow of chamber air (91.6 L min^{-1}) was measured during the 30 min by a flow meter (G/T 1000, Nr. 1024/MKC23BAL10000, Brooks Instruments B.V., Veenendaal, The Netherlands). Ammonia content of the outflowing air was analysed for NH_3 by a chemiluminescence analyser (model 17 & 42C; Thermo Environmental Instruments Inc., Franklin, MA, U.S.A.).

At the start and the end of the storage experiment, manures were thoroughly homogenised and samples were drawn. Total passive storage N losses were measured by the N balance technique (Pollet *et al.*, 1998). The ventilators enforced NH_3 emissions during the short periods of measurement leading to an average overestimation accounting for the 1.81 fold level of that of the unforced emission as determined by comparing NH_3 emission rates integrated over storage time and actual N losses according to N balances. Therefore, NH_3 data reflect potential losses rather than passive emissions during undisturbed storage. In contrast, integration over time yielded unbiased estimates of CH_4 and N_2O emissions. Greenhouse gas potential was calculated as kilogram-equivalents of CO_2 from the gaseous emissions measured using conversion factors of 21 and 310 for CH_4 and N_2O, respectively (IPCC, 1996). CO_2 emissions were not considered as CO_2 released from animals and manure in this case was only part of carbon cycling and no source of additional CO_2. This is different for additional CO_2 from fossile carbon sources combusted in conjunction with animal husbandry which was included in calculations for instance by Johnson *et al.* (2001). Carbon dioxide from fossile sources was not quantified here since it greatly differs even between farms using similar systems.

2.3. CHEMICAL ANALYSES

Dry matter and organic matter contents of feed and faeces were analysed by heating samples in a muffle furnace with steps at 105 and 550 °C using automatic weight loss measurement until weights remained constant (TGA 500, Leco Instruments, St. Joseph, MI, U.S.A.). Neutral detergent fibre (NDF) in feed and faeces was determined as recommended by Van Soest *et al.* (1991). Contents of fatty acids in feed were measured by gas chromatography (HP 5890A, Hewlett Packard, Avondale, PA, U.S.A.) equipped with a Supelcowax^{TM} 10 column (Fused Silica Capillary Column, 30 m × 0.32 mm) as described earlier (Dohme *et al.*, 2000). Nitrogen contents were analysed in feed, faeces and urine (automatic C/N-Analyzer type FP-2000, Leco instruments, St. Joseph, MI, U.S.A.) as well as manure (Kjeldahl technique). Initial manure contents of P and K were determined by inductively coupled plasma atomic emission spectroscopy (Perkin-Elmer Plasma II, Norfolk, CO, U.S.A.). Manure pH was measured with a pH meter (model 632 equipped with the electrode 6.0202.000 containing 3 M KCl electrolyte; Metrohm, Herisau, Switzerland) in homogenised samples within 30 min after mixing as well as at a depth of 10 cm at the start and the end of storage. These values corresponded well enough and were used to calculate initial and final pH. Manure carbon contents were determined by a CHN analyser (CHN-600; LECO Corporation, St. Joseph, MI, U.S.A.) in samples first dried at 80 °C for 20 hr and then at 105 °C for 2 hr.

2.4. Statistical evaluation

For the statistical evaluation of the results, data were subjected to analysis of variance by ANOVA applying the statistic package S+ (version 4, 1997; MathSoft Inc., Data Analysis Products Division, Seattle, WA, U.S.A.). Dietary treatment (α) was considered as main effect in the animal experiment (Equation (1)), while in the manure storage experiment as additional main effect manure type (β) as well as the interaction of diet × manure type ($\alpha\beta$) were regarded in Equation (2):

$$y_{ij} = \mu + \alpha_j + e_{ij} \tag{1}$$

$$y_{ijk} = \mu + \alpha_j + \beta_k + (\alpha\beta)_{jk} + e_{ijk} \; . \tag{2}$$

Furthermore, diet effects (α) within manure type were evaluated by monofactorial analyses of variance in Equation (3):

$$y_{ij} = \mu + \alpha_j + e_{ij} \; . \tag{3}$$

The tables and figures display mean values and the respective standard errors of the means. Complete slurry and the urine-rich slurry/farmyard manure system were compared using joint emission factors for urine-rich slurry and farmyard manure which were based on the figures related to the daily excretion of faeces and urine per cow also applying Equations (2) and (3), respectively.

3. Results

3.1. Dairy cow experiment

The complete rations as offered were similar in analysed nutrient composition, except their concentrations of C_{12} and C_{18} (Table I). The crude protein contents (6.25 × N) of 156 and 158 g kg^{-1} DM were within the range typical for winter

TABLE II

Differences between cow-related traits in the two dietary treatment groups[a]

Dietary treatment	C_{12}	C_{18}	SEM
Daily intake and performance			
DM (kg)	14.4* (13.9)	17.7** (17.9)	0.85
NDF[b] intake (kg)	5.1 (5.2)	5.8 (6.1)	0.31
N intake (g)	354* (309)	449** (442)	26.8
Milk yield[c] (kg)	27.0 (24.6)	28.0 (27.4)	1.51
Daily excretion			
Faeces (kg)	29.5 (29.6)	32.7 (32.8)	1.79
Urine (kg)	17.0 (13.9)	17.4 (13.9)	2.77
Complete faeces and urine (kg)	46.5 (43.5)	50.1 (46.6)	3.81
Faecal NDF[b] (kg)	2.4 (2.5)	2.4 (2.5)	0.13
Faecal N (g)	135* (125)	182** (181)	11.0
Urine N (g)	85 (63)	108 (89)	8.8
Complete faeces and urine N (g)	220* (188)	290** (270)	17.3
Urine N			
(% of total faeces and urine N)	38.2 (33.6)	37.1 (33.5)	1.74
NDF[b] content (g kg^{-1})			
Faeces			
In DM	522** (583)	430* (450)	17.4
As excreted	81.2 (85.0)	74.3 (77.0)	2.24
Complete faeces and urine			
As excreted	51.8 (57.7)	49.4 (54.3)	2.89
Methane release			
g day^{-1}	304* (304)	385** (403)	15.1
g kg^{-1} DM intake	21.3 (21.9)	21.9 (22.9)	1.08
g kg^{-1} digested NDF	112 (116)	118 (116)	7.81
g kg^{-1} milk	11.4* (12.3)	14.0** (15.0)	0.76

[a] Means by treatment, n = 6; values in brackets: means of cows whose manure was used for the storage experiment (n = 3). $*$, $**$, significant treatment differences at $P < 0.05$ applying Equation (1). SEM, standard error of mean.
[b] Neutral detergent fibre.
[c] Energy corrected milk after RAP (1999).

rations. The two complete diets also were equivalent in calculated supply of absorbable protein and net energy. Although cows of both dietary treatments were offered the same amount of feed, cows of the C_{12} group refused about half of the concentrate due to palatability reasons. This also changed the composition of the C_{12} diet as consumed. Particularly the forage to concentrate ratio was varied from 1.5:1 to 2.4:1. Also the respective calculated total dietary C_{12} and C_{18} contents (29 and 40 g kg^{-1}) and measured intakes (369 and 617 g day^{-1}) differed when the diets as consumed were compared. Table II gives the effects of the type of fatty acid in the diet and compares the overall means with those found for cows allocated to the manure storage experiment. These cows showed values very similar to the overall means in most traits, which justifies the use of this subpopulation that yielded the required manure at the same time point. There were certain diet effects. Concentrate refusals by the C_{12} group at almost unchanged forage intake caused a lower DM intake compared to the C_{18}-fed cows at a level of 3.3 kg d^{-1} ($P < 0.05$). This also reduced the intake of nitrogen and fibre (NDF). As only concentrate, containing less fibre than forage, was refused the NDF intake was relatively less affected by C_{12} than total feed intake. Considering certain initial differences in milk yield between the cows allocated to the C_{12} and C_{18} group (30.8 vs 28.7 kg d^{-1} milk; not given in Table II), performance was also reduced by C_{12}, which was more obvious in the selected subpopulation of cows (values in brackets) that did not differ in initial milk yield (27.6 vs 28.1 kg d^{-1} milk). The treatment differences in intake were reflected in excretion of faeces and nitrogen (significant for faecal N) while the proportion of urinary N in total N remained unaffected by the diet. C_{12}-fed cows digested NDF less efficiently than C_{18} cows (0.536 vs 0.576) and their faeces had a significantly higher NDF content in DM. Complete excreta were also slightly higher in NDF content in this group. Methane release (g day^{-1}) was significantly ($P < 0.05$) lower by 21% with C_{12} than with C_{18} but was equal per unit of fibre digested. The decline in methane release with C_{12} instead of C_{18} was 3 and 19%, respectively, when related to DM intake and milk yield, corresponding to the diet group differences in DM intake and milk yield.

3.2. Manure storage experiment

Manures differed in initial composition both with respect to dietary treatments and manure type according to the different excretory pattern of the animals and the deliberate variation between manure types in urine proportion (Table III). Manures of the C_{12}-fed cows had a significantly lower N content and, consequently, pH than manure from the C_{18} treatment while the C/N ratio did not differ. Similar to the variation in N content, the C_{12} manures had lower P contents than the C_{18} manures (0.58 vs 0.86 g kg^{-1}) but similar K contents (4.53 vs 4.70 g kg^{-1}). The initial group differences in total N and pH were still partly present after 14 weeks of storage, and small but partially significant differences were then found in the C/N ratio between dietary treatments. As the urine collected had been void of faeces

TABLE III

Effect of diet and manure type on manure composition and trace gas emission during manure storage[a]

	Type of manure									
	Complete slurry		Urine-rich slurry		Farmyard manure		Effect (P <) of			SEM
Type of diet	C_{12}	C_{18}	C_{12}	C_{18}	C_{12}	C_{18}	Diet	Manure	D × M	
Fresh manure										
Total N (g kg^{-1} wet weight)	4.57**	6.80*	12.5**	16.1*	5.03**	6.26*	0.001	0.001	0.001	0.052
pH	6.32**	6.67*	6.79**	7.50*	6.96	6.35	ns	0.05	0.05	0.468
C/N ratio[b]	18.9	18.4	17.5	17.4	24.7	23.5	–	–	–	–
Manure stored for 14 weeks										
Total N (g kg^{-1} wet weight)	3.11**	4.89*	2.73	2.71	4.06**	4.82*	0.001	0.001	0.01	0.429
pH	9.08	9.06	9.74	9.69	8.84	8.43	ns	0.001	ns	0.408
C/N ratio	20.9*	20.2**	17.8	16.8	17.2**	17.3*	ns	0.001	ns	1.72
Average emission rate (mg kg^{-1} undiluted manure d^{-1}) after 10 days of storage										
CH_4	0.07**	3.81*	0.09**	0.84*	24.82**	29.40*	ns	0.001	ns	4.414
Average emission rate (mg kg^{-1} undiluted manure d^{-1}) over 14 weeks of storage										
CH_4	29.06*	8.90**	7.09	2.62	10.76*	8.90**	0.01	0.01	0.05	5.371
N_2O	0.86	0.96	0.02	0.03	1.34	2.99	0.01	0.001	0.01	0.430
NH_3	47.7	54.3	63.9**	91.9*	30.7	43.7	0.001	0.001	0.05	8.18
Total nitrogen loss over 14 weeks of storage										
% of initial	32.9	31.0	45.1	48.0	14.1	15.8	ns	0.001	ns	8.08
g cow^{-1} d^{-1}	62.1**	88.6*	85.3**	137.3*	26.7**	45.1*	0.001	0.001	0.01	9.08

[a] Means by subgroup, n = 4. *,** Means with unequal superscripts within manure type are significantly different at $P < 0.05$ according to statistical Equation (3). Probability of error (P) for significance of effects describe results of Equation (2). D × M, interaction of diet and type of manure. SEM, standard error of mean (Equation (2)). [b] Not statistically tested since only one determination per group available.

particles, spontaneous N loss from non-acidified urine during collection was found to be very low in occasional control measurements ensuring that samples used for manure mixing were still representative with respect to initial N content.

Methane release from the C_{12} and C_{18} treatment manures showed a different evolution with storage time (Figure 1). Methane emission started earlier with C_{18} in the complete slurry, with significantly greater values at day 10 of storage as one deliberately selected time point reflecting an early stage of storage (Table III). This was later reversed leading to overall higher CH_4 emission rates in all manure types derived from the C_{12}-fed cows compared with the C_{18} manures (significant for complete slurry and farmyard manure). In the case of complete slurry derived from the C_{12}-fed cows, the maximum of CH_4 emission was only reached shortly before the end of the 14 weeks lasting storage period (Figure 1). When related to the faeces and urine amount as daily excreted by the cows, the diet effects were even more clearly expressed in complete slurry after 14 weeks of storage although effects were much less pronounced after 7 weeks of storage (Table IV). In the urine-rich slurry/farmyard manure system, CH_4 emissions were lower during 14 weeks of storage than in slurry. The pattern of CH_4 release with time also clearly differed between manure types with high initial values in farmyard manure and a retarded onset of emission in the slurries (Figure 1).

Combining ruminal and manure emissions over 14 weeks of storage, CH_4 release was lower by 26 g cow^{-1} day^{-1} (5.8%) with C_{12} than with C_{18} in the complete slurry system (Table IV). The corresponding reduction was 90 g cow^{-1} day^{-1} (21.3%) in the two-component system. Accordingly, the reduction found in animal-derived CH_4 release was mostly compensated by manure CH_4 in complete slurry and partially in the two-component system. The proportions of total CH_4 emission originating from manure storage over 7 weeks were 7.8 and 8.7% in the slurry system and the two-component system, respectively. The corresponding values for 14 weeks of storage were 18.4 and 12.5%. Total CH_4 released per unit of milk combining data of both storage systems was lower with C_{12} than with C_{18} applying a storage period of 7 weeks (13.4 vs 14.1 g kg^{-1}; $P < 0.05$) whereas the ranking of the average values was reversed after 14 weeks of storage (15.3 vs 14.4 g kg^{-1}).

Emissions of N_2O were low in urine-rich slurry and remained unaffected by the diet. Diet also had no effect on N_2O emissions from the complete slurry, but complete slurry had a N_2O emission rate 43 times higher than the urine-rich slurry (Table III). The level of N_2O emission was highest in farmyard manure, particularly from the C_{18}-fed cows, which reached a level of N_2O more than twice as high as in the C_{12} manure. The onset of significant N_2O emissions in complete slurry and farmyard manure took place not before 7 or 8 weeks of storage had passed (Figure 2). When related to the daily amount of excreta per cow, N_2O emissions were similar in all treatments except in the urine-rich slurry/farmyard manure system with C_{18} slurry (Table IV). Overall N_2O emissions partially counterbalanced CH_4 emissions from manure in terms of greenhouse gas potential and corroborated with treatment differences in animal CH_4 release. Total greenhouse equivalents

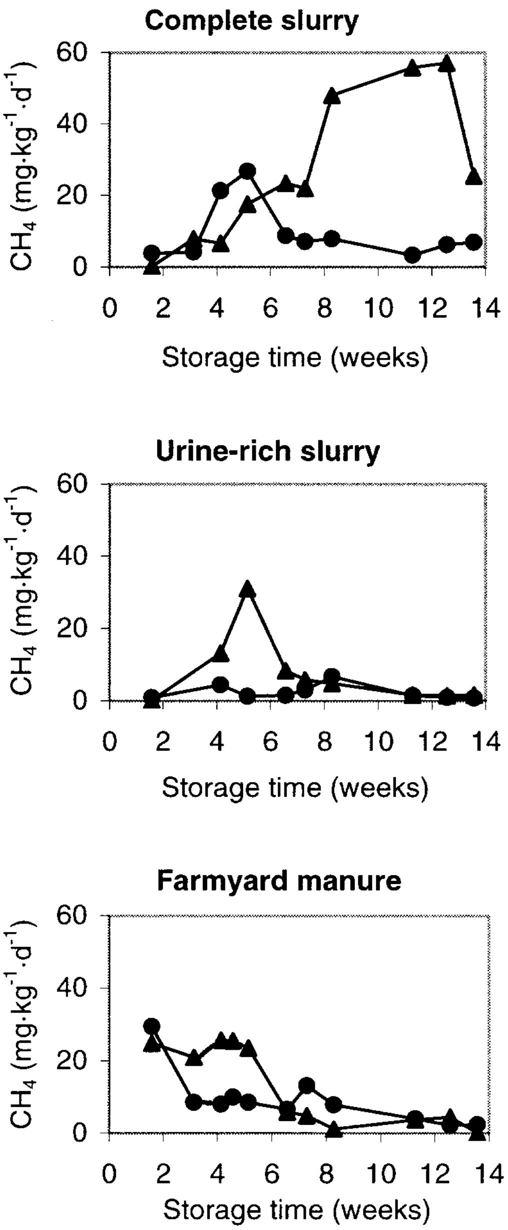

Figure 1. Effects of diet and manure type on the evolution of CH_4 emission rate during manure storage (▲, C_{12} diet; ●, C_{18} diet). Each value reflects the average of twelve individual measurements (four replicated buckets and three extracted air samples per time point and bucket).

TABLE IV

Total emission of greenhouse gases via animal and stored manure related to the daily amount of faeces and urine per cow[a]

	Type of manure							
	Complete slurry		Urine-rich slurry and farmyard manure		Effect (P <) of			SEM
Type of diet	C_{12}	C_{18}	C_{12}	C_{18}	Diet	Manure	D × M	
Methane (g cow^{-1} d^{-1})								
Manure (7 weeks of storage)	31.0	27.1	20.4	10.9	0.05	ns	0.05	8.95
Manure (14 weeks of storage)	115.3*	42.2**	27.6	18.8	0.001	0.001	0.001	14.16
Cow[b] and manure (7 weeks of storage)	335.0**	429.6*	324.4**	413.4*	0.001	ns	0.05	8.95
Cow[b] and manure (14 weeks of storage)	419.3	445.0	331.6	421.6	0.001	0.001	0.001	14.16
Methane originating from manure (% of total)								
7 weeks of storage	9.2	6.3	12.2	5.2	0.01	ns	0.05	2.231
14 weeks of storage	27.3*	9.5**	16.2*	8.7**	0.001	0.001	0.001	2.883
Nitrous oxide (g cow^{-1} d^{-1})								
Manure (7 weeks of storage)	0.06	0.05	0.99	1.49	ns	0.01	ns	1.207
Manure (14 weeks of storage)	3.67	4.41	1.88	4.74	0.01	0.01	0.05	1.768
Greenhouse gas equivalents of CO_2 (kg cow^{-1} d^{-1})[c]								
7 weeks of manure storage	7.05**	9.04*	7.12**	9.14*	0.001	0.01	ns	0.491
14 weeks of manure storage	9.94	10.71	7.55	10.32	0.001	ns	0.01	0.705

[a] Means by subgroup, n = 4. *,** Means with unequal superscripts within manure type are significantly different at $P < 0.05$ according to statistical Equation (2). Probability of error (P) for significance of effects describe results of Equation (2). D × M, interaction of diet and type of manure. SEM, standard error of mean (Equation (2)).
[b] 304.0 and 402.5 g methane cow^{-1} d^{-1}; data from Table II.
[c] Considering animal and manure-derived CH_4 as well as N_2O from manure.

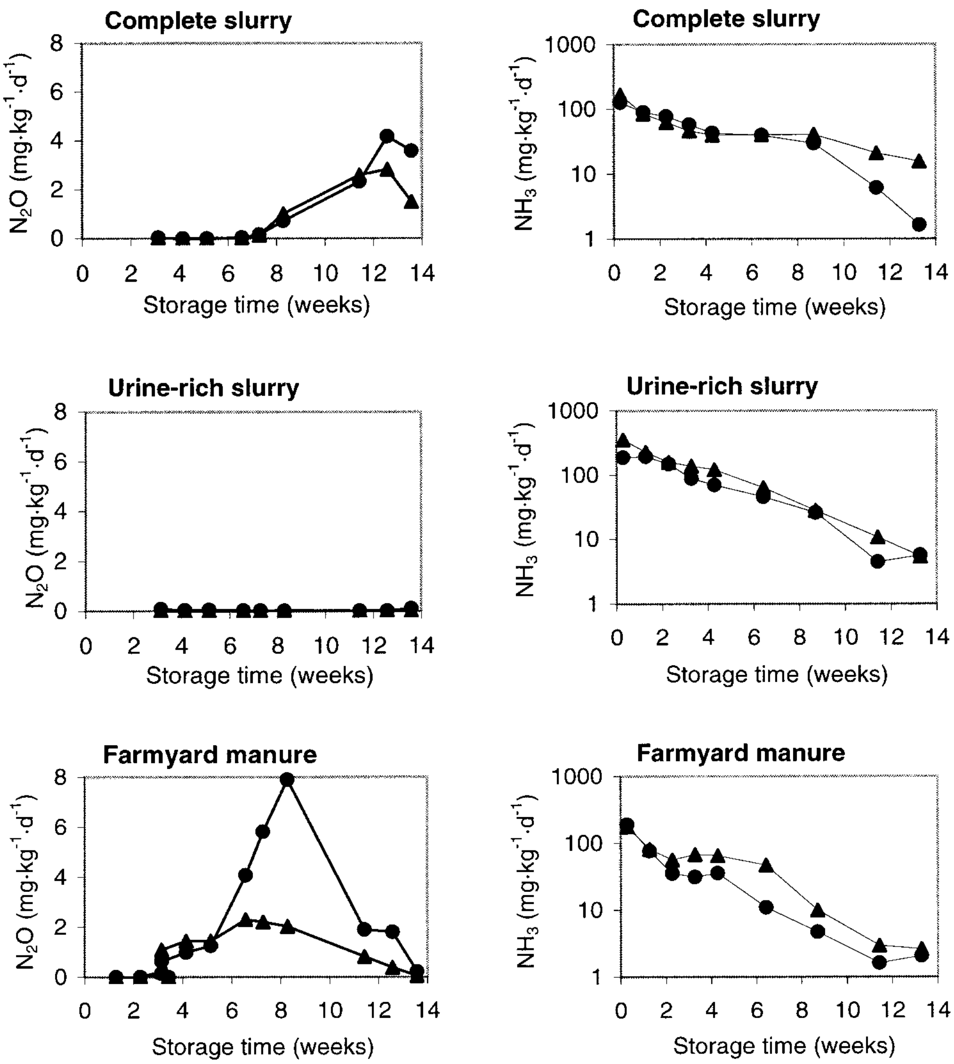

Figure 2. Effects of diet and manure type on the evolution of N_2O (left) and NH_3 (right) emission rate during manure storage (▲, C_{12} diet; ●, C_{18} diet). Each N_2O (three extracted air samples per time point and bucket) and NH_3 value reflects the average of twelve and four individual measurements (four replicated buckets per value), respectively.

(GHGeq) from CH_4 (including animal-derived CH_4) and N_2O were lower in the C_{12} treatment than with C_{18} (Table IV). Differences between types of slurry were less clear. Nitrous oxide contributed 0.2 and 12.1% to the GHGeq in the system with complete slurry stored for 7 and 14 weeks, respectively. The corresponding values for the two-component manure storage system were 4.7 and 11.5%. Overall, N_2O emissions made up 51% of manure-derived GHGeq (31 to 79%) during 14 weeks of manure storage.

Total N losses and NH_3 emission rates over 14 weeks of storage were generally higher in the C_{18} than in the C_{12} treatment in all manure types ($P < 0.05$), while the losses relative to initial manure N contents were quite similar between dietary treatments (Table III). Ammonia emission showed a logarithmically-shaped decline without major dietary differences, except in complete slurry when after 7 weeks of storage a pronounced NH_3 decrease in the C_{18} but not in the C_{12} treatment occurred (Figure 2).

4. Discussion

4.1. Effect of dietary lipid source on animal and manure-derived methane emission

In ruminant husbandry, CH_4 formation is mainly the result of fibre fermentation both in the rumen and, at a proportion of 6 to 14 %, in the hindgut (Immig, 1996). Easily-degradable nutrients such as starch contribute far less to CH_4 release (Kreuzer *et al.*, 1986). This may explain the almost identical CH_4 rates per unit of dry matter intake and, associated with that, digested fibre found in the two groups within the present study. Lipids have been identified as effective nutritional options to suppress ruminal CH_4 formation (Van Nevel and Demeyer, 1996; Moss *et al.*, 2000). Focus was first put on oils rich in polyunsaturated fatty acids. However, these fatty acids have a strong negative influence on fibre degradation in the rumen (Van Nevel and Demeyer, 1996), and so their use is associated with a considerable loss of energy. More recently, the efficacy of various fats rich in saturated fatty acids of medium chain length against methane release was shown *in vitro* (Dohme *et al.*, 2000) and *in vivo* (Machmüller and Kreuzer, 1999; Machmüller *et al.*, 2000). These fats affect fibre digestion in the rumen and the total digestive tract to a smaller extent than unsaturated oils, as can also be seen from the present study with only an insignificant decrease in fibre digestibility using C_{12} instead of C_{18}. Dohme *et al.* (2001) identified C_{12} and C_{14} (myristic acid) *in vitro* as the only members of the group of the medium-chain fatty acids (C_8 to C_{16}) active against ruminal CH_4 formation. However, in the present study C_{12} failed to exhibit a clear CH_4 suppressing effect when expressed per kg DM intake. The lower CH_4 release of the C_{12}-fed cows compared with those receiving C_{18} could be explained by the associated suppression of concentrate intake to about half of the amount allocated. This was presumably a result of the particular odour and soapy taste of C_{12}. The reasons for the lack of a direct CH_4 suppressing effect of C_{12} remain unclear. The significant refusal of concentrate, associated with a higher dietary proportion of forage, may have decreased passage rate through the rumen enabling methanogens to produce more CH_4 than at an unchanged forage to concentrate ratio. Also, interactions with carbohydrates and minerals (Soliva *et al.*, 2000), binding and inactivating particularly free C_{12}, may explain why the *in* *vivo* efficacy was higher with the use of fats such as coconut oil where the C_{12} was incorporated in triglycerides (Machmüller and Kreuzer, 1999; Machmüller *et al.*, 2000). Another reason could be a compensation of a lower ruminal fibre digestion and CH_4 formation by a correspondingly increased activity of the hindgut, which was noted for fibre digestion with coconut oil supplementation before (Sutton *et al.*, 1983). The potential CH_4-inhibiting action of C_{12} is no longer present in the hindgut, since at this site C_{12} has been already absorbed from the gut. However, the rate of CH_4 released per unit of fermented fibre is also lower in the hindgut than in the rumen (Immig, 1996).

The use of C_{12} increased the fibre content of the faeces in the present study because the forage proportion in feed was increased and fibre digestibility was slightly decreased. Nevertheless, the amount of fibre (NDF) excreted per cow per day was identical in both groups. Accordingly, summarising 7 weeks of manure storage, complete slurry showed no diet effect on CH_4 emission from manure when related to daily excreta amount per cow. However, CH_4 emission was higher with C_{12} in the urine-rich slurry/farmyard manure system stored for 7 and 14 weeks and in complete slurry stored for 14 weeks. It appears that the compositional differences between the C_{12} and the C_{18} type of manure resulted in a higher CH_4 release only under favourable fermentation conditions, which seems to have been achieved earlier in the two-component system than in the complete slurry system (cf. Figure 1). The known antagonism of NH_3 and CH_4 release from manure (Hansen *et al.*, 1998) does not give an explanation for the retarded onset of the major part of CH_4 release only in the C_{12} group because NH_3 declined even more rapidly in the C_{18} type of manure. When manure was only stored for 7 weeks, feeding of C_{12} instead of C_{18} generally resulted in slightly lower overall (animal together with manure) CH_4 emission per kg of milk which is an important measure of efficiency. This effect disappeared with 14 weeks of storage, and even the positive effect found at 7 weeks of storage duration is questionable since the C_{12}-fed cows performed too well in terms of milk yield regarding their reduced feed intake, so being obviously forced to mobilise significant quantities of energy from body reserves. In comparison with the C_{18} group, the C_{12}-fed cows later have to compensate for this spent energy (approx. 12 MJ day^{-1}) by a correspondingly higher DM intake (1.7 kg day^{-1}) equivalent to daily 44 g enteric CH_4. This is half of the difference of the 81 g enteric CH_4 day^{-1} found between the C_{12} and the C_{18} group.

4.2. Effect of Type and Duration of Manure Storage on Methane Release

Values on CH_4 emission from differently stored manure are still scarce. According to the present results, the levels and the differences between manure storage systems in CH_4 emissions were distinctly determined by the duration of storage. Initially farmyard manure was a more important source of CH_4 than the two types of slurry similar to previous findings (Külling *et al.*, 2001, 2002), with higher values in the urine-rich slurry/farmyard manure system than with complete slurry. However, the ranking changed around week 7, when CH_4 emission had almost ceased in all manures, except in complete slurry. Over a storage period of 14 weeks, CH_4 emissions from the urine-rich slurry/farmyard manure system were lower than for complete slurry. Two thirds of the CH_4 emission from solid manure storage were emitted within the first of three months in experiments of Amon *et al.* (1998, 2001). With farmyard manure also in the present study the major proportion of CH_4 was released within this period, but not with complete slurry. Undisturbed crust formation and surface covers may substantially reduce CH_4 release from slurry, and the onset of the major CH_4 release can be retarded (Sommer *et al.*, 2000). Accordingly, in order to get reliable data for a feeding or manure system comparison in CH_4 emission, the measurement period should be chosen sufficiently long, particularly with slurry systems.

Decomposition of animal manures is potentially a larger source of CH_4 than fermentation in the gastrointestinal tract provided disposals are primarily subjected to anaerobic conditions (Johnson and Ward, 1996). However, in the present study manures only contributed between 5 and 27% (11.8% on average) to total CH_4 emission. This was in the range of 13 to 14% assumed for various calculations (Anonymous, 1996 and 1999; Minonzio *et al.*, 1998). The value is higher with 14 weeks (15%) and lower with 7 weeks of storage (8%) then being comparable to data from Kinsman *et al.* (1995) of about 6%.

Methane release from manure almost exclusively takes place during storage and no longer at the aerobic conditions after field distribution (Yamulki *et al.*, 1999). Therefore, it can be assumed that the CH_4 emission as reported here comprises almost the complete CH_4 loss from dairy husbandry under the conditions described. However, the present results only represent a certain stage of the lactation cycle of the cows. In order to be able to give annual data, standard milk yield curves and/or feeding intensity level have to be considered (Pelchen *et al.*, 1998). Kirchgessner *et al.* (1991) developed regressions on annual CH_4 emission of cows based on lactational milk yields. Accordingly, from the present data annual CH_4 emission rates of the C_{18}-fed cows (controls; estimated annual milk yield of 7000 kg) can be calculated to account for approximately 120 kg cow^{-1} a^{-1}. These values have to be adjusted by the additional CH_4 from manure adding up to 130 and 138 kg CH_4 cow^{-1} a^{-1} for 7 and 14 weeks of manure storage, respectively. This is slightly above the range of other studies of 117 to 127 kg cow^{-1} a^{-1} (Kinsman *et al.*, 1995; Sneath *et al.*, 1997, Seipelt *et al.*, 1999) using cows of similar milk yield (Kinsman *et al.*, 1995) as in the present investigation. In contrast the estimate of Amon *et al.* (1998) is far lower with 71 kg cow^{-1} a^{-1} which can be explained by the presumably lower feed intake of the cows investigated as they only produced 18.5 kg milk day^{-1} (equivalent to approx. 4500 kg milk a^{-1}). Estimates on yearly CH_4 formation are similar for the C_{12} treatment when applying 14 weeks of storage and slightly lower with 7 weeks of manure storage. The manure-derived CH_4 from the C_{18}-fed cows, adjusted to the actual Swiss situation with 70% of dairy excreta stored in the urine-rich slurry/farmyard manure system (Menzi *et al.*, 1997), can be estimated to account for 5.8 and 9.4 kg cow^{-1} a^{-1} with 7 and 14 weeks of storage, respectively. The value measured for 14 weeks of storage corresponds well to the value of 10.5 kg cow^{-1} a^{-1} applied for the Swiss national CH_4 inventory (Minonzio *et al.*, 1998), but is lower than the IPCC standard value of 14.0 kg cow^{-1} a^{-1} (Anonymous, 1996).

4.3. Effect of type of diet and manure on nitrogenous emissions

Nitrogen emissions were quantified in the present study as total gaseous N loss and as NH_3 as well as N_2O. In line with the lower N intake and N excretion of the C_{12}-fed cows, total N and NH_3 emission during storage were lower in the C_{12} treatment than in the C_{18} treatment in all types of manure, and N losses relative to initial N content remained unaffected. This was expected since the proportion of urine N, which is rapidly converted to ammoniacal N in manure, was not varied by the type of diet. The crude protein content of the diet, as one major factor determining urine N percentage of total N (Külling *et al.*, 2001), was unchanged between diets. Nitrous oxide, by contrast, was clearly lower in the manures from the C_{12} treatment. One reason for this observation might be sought in the antagonism of N_2O and NH_3 release, which showed a contrary development in terms of level and evolution in the different treatment groups. In the experiments of Külling *et al.* (2001, 2002) N_2O level was also found to be low as long as NH_3 emission did not reach a low value. Similar to composting of household waste (Beck-Friis *et al.*, 2000), N_2O thus considerably increased with a prolonged storage of manure.

The system comparison of complete slurry and the two-component type of storage overall demonstrated quite similar total N losses (75 vs 74 g cow^{-1} d^{-1}), NH_3 emission rates (60 vs 68 μg m^{-2} s^{-1}) and N_2O emissions (4.0 vs 3.3 g cow^{-1} d^{-1}) during 14 weeks of storage. However, during 7 weeks of storage, N_2O emissions were 23 times higher in the urine-rich slurry/farmyard manure system than with complete slurry. Overall, these N-related effects of storage type confirm results of an earlier system comparison investigating a wide range of dietary crude protein contents either through modifications in forage (Külling *et al.*, 2002) or in concentrate (Külling *et al.*, 2001). In the two-component manure storage system, N_2O originated almost exclusively from farmyard manure as expected while in urine-rich slurry N_2O was mostly not detectable in the air samples extracted from the closed chambers. Extrapolated to an annual basis, emissions were 0.2 and 1.3 kg cow^{-1} a^{-1} of N_2O within 7 and 14 weeks of storage, respectively, on average of all manure types and dietary treatments. The level of N_2O emitted within 7 weeks was lower than the 0.7 kg cow^{-1} a^{-1} found previously during the same period of storage (Külling *et al.*, 2001), whereas the amount calculated for 14 weeks of storage was quite similar to the value of 1.6 kg cow^{-1} a^{-1} estimated by Schmid *et al.* (2000).

Different from NH_3, where emission rapidly declines with time, for reliable measurements of total N_2O release like with CH_4 an extended storage duration turned out to be very important. Experimentally restricting storage to periods shorter than actually applied in farm practice thus may lead to both an underestimation of the real level and misleading results of diet and manure system comparisons.

4.4. Effect of type of diet and manure on global warming potential of dairy husbandry

There was a certain compensation of differences between diets and between manure systems (after 14 weeks of storage) in overall CH_4 and in N_2O emissions in terms of overall global warming potential by the greenhouse gases due to their mostly contrary variation with treatment. Despite its low absolute levels, N_2O contributed about 7% to total and 51% to manure-derived GHGeq after 14 weeks of manure storage owing to its far higher global warming potential compared with CH_4. However, at a reduced storage time of 7 weeks, N_2O proportion of total GHGeq was minor in the two-component system and negligible in complete slurry. By contrast, N_2O was estimated to be more important for GHGeq than CH_4 in U.S. dairy (1.5 fold; Phetteplace *et al.*, 2001) and beef production systems (1.2 fold; Johnson *et al.*, 2001). However, these studies assumed high N_2O losses during grazing on well fertilised pastures, and CH_4 emission from excreta is low under pasture conditions (Williams, 1993). Furthermore manure-derived N_2O is only one source of total direct and indirect system N_2O losses.

5. Conclusions

In this study for one of the first times a direct comparison of CH_4 emitted from animals and manure was carried out within one investigation. The results indicate that there could be a certain tendency for a compensatory increase in CH_4 formation in manure in cases of reduced enteric CH_4 formation when associated with a reduced fibre fermentation. It is nevertheless considered profitable to reduce enteric CH_4 formation since this CH_4 is inevitably lost whereas manure storage technology and duration offer opportunities to avoid high CH_4 losses or even allow to collect CH_4 as biogas. It still remains open whether or not a compensation takes place in situations of a CH_4 suppression independent from digestible fibre supply. The relatively long manure storage duration followed and the consideration of ruminal CH_4 emissions gave new insights on long-term effects of feeding and manure type. Irrespective of manure type and storage duration, enteric CH_4 release obviously is always of greater importance for the global warming potential of dairy husbandry systems than CH_4 and N_2O emissions during manure storage, but the contribution of these two trace gases for GHGeq increases with storage time. Methane emissions therefore have to be considered in a more integrated and holistic way in future as has been recommended and partly realised earlier (Demeyer and Van Cleemput, 1996). Also further assessment of the relative importance of CH_4 and N_2O for GHGeq under various dietary conditions and on pasture is necessary.

Acknowledgements

We are grateful to Dr. H. Leuenberger, Dr. A. Neftel, A. Felder and M. Schmid for their assistance in organising and performing the investigations at the ETH Research Station Chamau and at the Federal Research Station of Agroecology and Agriculture (FAL), Liebefeld. We also owe thanks for assistance in the laboratory to H. Shariat-Madari, T. Probst and J. Aeberhard.

References

Agnihotri, S., Kulshreshtha, K. and Singh, S. N.: 1999, 'Mitigation strategy to contain methane emission from rice-fields', *Environ. Monit. Assess.* **58**(1), 95–104.

Amon, B., Boxberger, J. and Amon, T.: 1998, 'Untersuchung der Ammoniakemission in der Landwirtschaft Österreichs zur Ermittlung der Reduktionspotentiale und Reduktionsmöglichkeiten', *Endbericht zum Forschungsprojekt, Nr. L 883/94*, Institut für Land-, Umwelt- und Energietechnik, Universität für Bodenkultur, Vienna, Austria.

Amon, B., Amon, T. and Boxberger, J.: 2001, 'Emissions of NH_3, N_2O and CH_4 from dairy cows housed in a farmyard manure tying stall (housing, manure storage, manure spreading', *Nutr. Cycl. Agroecosyst.* **60**(1/3), 103–113.

Anonymous: 1996, *IPCC Guidelines for National Greenhouse Gas Inventories*, 3. IPCC/OECD/IEA, U.K. Meteorological Office, Bracknell, U.K.

Anonymous: 1999, *Annual European Community Greenhouse Gas Inventory 1990–1996*, Submission from 21 April 1999 to the Secretariat of the UNFCCC, European Commission DG XI Environment, Nuclear Safety and Civil Protection, European Environment Agency, Copenhagen, Denmark.

Beck-Friis, B., Pell, M., Sonesson, U., Jönsson, H. and Kirchmann, H.: 2000, 'Formation and emission of N_2O and CH_4 from compost heaps of organic household waste', *Environ. Monit. Assess.* **62**(3), 317–331.

Chynoweth, D. P.: 1996, 'Environmental impact of biomethanogenesis', *Environ. Monit. Assess.* **42**(1–2), 3–18.

Demeyer, D. I. and Van Cleemput, O. (eds): 1996, 'Methane emissions through animals and from the ground', Special Issue, *Environ. Monit. Assess.* **42**(1–2), 1–210.

Dohme, F., Machmüller, A., Wasserfallen, A. and Kreuzer, M.: 2000, 'Comparative efficiency of various fats rich in medium-chain fatty acids to suppress ruminal methanogenesis as measured with Rusitec', *Can. J. Anim. Sci.* **80**(3), 473–482.

Dohme, F., Machmüller, A., Wasserfallen, A. and Kreuzer, M.: 2001, 'Ruminal methanogenesis as influenced by individual fatty acids supplemented to complete ruminant diets', *Lett. Appl. Microbiol.* **32**(1), 47–51.

Hansen, K. H., Angelidaki, I. and Ahring, B. K.: 1998, 'Anaerobic digestion of swine manure: Inhibition by ammonia', *Water Res.* **32**(1), 5–12.

Hashimoto, A. G., Varel, V. H. and Chen, Y. R.: 1981, 'Ultimate methane yield from beef cattle manure: Effect of temperature, ration instruments, antibiotics and manure age', *Agric. Wastes* **3**, 241–256.

Immig, I.: 1996, 'The rumen and hindgut as source of ruminant methanogenesis', *Environ. Monit. Assess.* **42**(1–2), 57–72.

IPCC (Intergovernmental Panel on Climate Change): 1996, in *Climate Change 1995 – The Science of Climate Change*, J. T. Houghton, G. J. Jenkins and J. J. Ephraums (eds), Cambridge University Press, U.K.

Johnson, D. E. and Ward, G. M.: 1996, 'Estimates of animal methane emissions', *Environ. Monit. Assess.* **42**(1–2), 133–141.

Johnson, D. E., Seidl, A. F. and Phetteplace, H. W.: 2001, 'Methane, Nitrous Oxide and Carbon Dioxide Emissions from U.S. Beef Production Systems', in A. Chwalibog and K. Jakobsen (eds), *Energy Metabolism in Animals* EAAP Publ. 103, Wageningen Pers, Wageningen, The Netherlands, pp. 161–164.

Kinsman, R., Sauer, F. D., Jackson, H. A. and Wolynetz, M. S.: 1995, 'Methane and carbon dioxide emissions from dairy cows in full lactation monitored over a six-month period', *J. Dairy Sci.* **78**(12), 2760–2766.

Kirchgessner, M., Windisch, W., Müller, H. L. and Kreuzer, M.: 1991, 'Release of methane and of carbon dioxide by dairy cattle', *Agribiol. Res.* **44**(2–3), 91–102.

Kreuzer, M., Kirchgessner, M. and Müller, H. L.: 1986, 'Effect of defaunation on the loss of energy in wethers fed different quantities of cellulose and normal or steamflaked maize starch', *Anim. Feed Sci. Technol.* **16**, 233–241.

Külling, D. R., Menzi, H., Kröber, T. F., Neftel, A., Sutter, F., Lischer, P. and Kreuzer, M.: 2001, 'Emissions of ammonia, nitrous oxide and methane from different types of dairy manure during storage as affected by dietary protein content', *J. Agric. Sci. Camb.* **137**(5), 235–250.

Külling, D. R., Menzi, H., Sutter, F., Lischer, P. and Kreuzer, M.: 2002, 'Ammonia, nitrous oxide and methane emissions from differently stored dairy manure derived from grass- and hay-based rations', *Nutr. Cycl. Agroecosyst*. (in press).

Machmüller, A. and Kreuzer, M.: 1999, 'Methane suppression by coconut oil and associated effects on nutrient and energy balance in sheep', *Can. J. Anim. Sci.* **79**(1), 65–72.

Machmüller, A., Ossowski, D. A. and Kreuzer, M.: 2000, 'Comparative evaluation of the effects of coconut oil, oilseeds and crystalline fat on methane release, digestion and energy balance in lambs', *Anim. Feed Sci. Technol.* **85**(1–2), 41–60.

Menzi, H., Frick, R. and Kaufmann, R.: 1997, 'Ammoniak-Emissionen in der Schweiz: Ausmass und technische Beurteilung des Reduktionspotentials', *Schriftenreihe der FAL*, 26, Eidgenössische Forschungsanstalt für Agrarökologie und Landbau, Zürich-Reckenholz, Switzerland.

Minonzio, G., Grub, A. and Fuhrer, J.: 1998, 'Methan-Emissionen der Schweizerischen Landwirtschaft', *Schriftenreihe Umwelt*, 298, Bundesamt für Umwelt, Wald und Landschaft (BUWAL) Berne, Switzerland.

Mosier, A. R.: 1989, 'Chamber and Isotope Techniques', in M. O. Andreae and D. S. Schimel (eds), *Exchange of Trace Gases between Terrestrial Ecosystems and the Atmosphere*, New York, John Wiley & Sons, pp. 175–187.

Moss, A. R., Jouany, J.-P. and Newbold, J.: 2000, 'Methane production in ruminants: its contribution to global warming', *Ann. Zootech.* **49**(3), 231–253.

Pelchen, A., Peters, K. J. and Holter, J. B.: 1998, 'Prediction of methane emissions from lactating dairy cows', *Arch. Anim. Breed.* **41**(6), 553–563.

Phetteplace, H. W., Johnson, D. E. and Seidl, A.: 2001, 'Greenhouse gas emissions from simulated beef and dairy livestock systems in the United States', *Nutr. Cycl. Agroecosyst.* **60**(1/3), 99–102.

Pollet, I., Christiaens, J. and Van Langenhove, H.: 1998, 'Determination of the ammonia emission from cubicle houses for dairy cows based on a mass balance', *J. Agric. Engin. Res.* **71**(3), 239–248.

RAP (Station fédérale de recherches en production animale): 1999, 'Fütterungsempfehlungen und Nährwerttabellen für Wiederkäuer', 4th ed., Landwirtschaftliche Lehrmittelzentrale, Zollikofen, Switzerland.

Schmid, M., Neftel, A. and Fuhrer, J.: 2000, 'Lachgasemissionen aus der Schweizer Landwirtschaft', *Schriftenreihe der Eidgenössischen Forschungsanstalt für Agrarökologie und Landbau (FAL)* 33, 131 pp., FAL Zürich-Reckenholz, Switzerland.

Seipelt, F., Ross, A. Steffens, G. and Van den Weghe, H.: 1999, Quantifizierung gasförmiger Emissionen aus frei gelüfteten Milchviehställen mittels Tracergaseinsatz nach der Abklingmethode', in *Construction, Engineering and Environment in Livestock Farming*, Freising-Weihenstephan, Germany, pp. 69–74.

Sneath, R. W., Phillips, V. R., Demmers, T. G. M., Burgess, L. R., Short, J. L. and Welch, S. K.: 1997, 'Long Term Measurements of Greenhouse Gas Emissions from U.K. Livestock Buildings', in *Livestock Environment V*, Bloomington, Minnesota, U.S.A., pp. 146–153.

Soliva, C. R., Kreuzer, M. and Machmüller, A.: 2000, 'Methane suppressing effect of lauric acid in rumen as influenced by carbohydrate composition and calcium', *Nutr. Reprod. Develop.* **40**(2), 212.

Sommer, S. G., Petersen, S. O. and Søgaard, H. T.: 2000, 'Greenhouse gas emission from stored livestock slurry', *J. Environm. Qual.* **29**(3), 744–750.

Sutter, F. and Beever, D. E.: 2000, 'Energy and nitrogen metabolism in Holstein-Friesian cows during early lactation', *Anim. Sci.* **70**(3), 503–514.

Sutton, J. D., Knight, R., McAllan, A. B. and Smith, R. H.: 1983, 'Digestion and synthesis in the rumen of sheep given diets supplemented with free and protected oils', *Br. J. Nutr.* **49**, 419–432.

Van Nevel, C. J. and Demeyer, D. I.: 1996, 'Control of rumen methanogenesis', *Environ. Monit. Assess.* **42**(1–2), 73–97.

Van Soest, P. J., Robertson, J. B. and Lewis, B. A.: 1991, 'Methods for dietary fiber, neutral detergent fiber, and nonstarch polysaccharides in relation to animal nutrition', *J. Dairy Sci.* **74**(10), 3583–3597.

Walther, U., Menzi, H., Ryser, J.-P., Flisch, R., Jeangros, B., Kessler, W., Maillard, A., Siegenthaler, A. F. and Vuillod, P. A.: 1994, 'Grundlagen für die Düngung im Acker- und Futterbau', *Agrarforschung* **1**(7), Supplement, 40 pp.

Williams, D. J.: 1993, 'Methane emissions from manure of free-range dairy cows', *Chemosphere* **26**(1–4), 179–187.

Yamulki, S., Jarvis, S. C. and Owen, P. (1999), 'Methane emission and uptake from soils as influenced by excreta deposition from grazing animals', *J. Environ. Qual.* **28**(2), 676–682.